MATHEMATICAL MODEL OF HETEROGENEOUS REACTIONS

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We study theoretically the chemical transformation of spherical particles. Equations are presented which describe the time dependence of the degree of chemical transformation of the original solid reagent and the reaction rate.

We consider the reaction of a spherical porous particle with gas or liquid in a sufficiently general form, but limit ourselves to reactions which do not change the volume of the particle.

The macrokinetics of the reaction will be represented as follows. The gaseous or liquid reagent reaches the surface of the particle by convection. Subsequently, it diffuses in the bulk of the solid reagent and simultaneously reacts with it. The gaseous or liquid product which is thus formed escapes from the reaction zone by diffusion and mass exchange into the surrounding medium. In the course of the reaction, we neglect the change of volume of the fluid reagent and also the kinetic and diffusion characteristics of the solid. At any point the reaction proceeds until all reagent at this point is used up. As a result, starting from a definite moment of time, a crust of the solid reaction product is formed on the surface of the particle. The thickness of this crust increases as the reaction develops until all volume of the particle undergoes the reaction.

In the course of the reaction, the structure of the interior of the particle can change qualitatively and, as a result, the diffusion coefficient of the gaseous reagent in the solid product can differ from its value in the original solid reagent. In each concrete case, the structure of the particle can change in a specific manner which is difficult to describe analytically. Therefore, to simplify the problem and obtain quantitative estimates of the effect of the change in the diffusion constant, we have, in the first approximation, assumed that the diffusion coefficient remains constant in the whole reaction zone, and changes stepwise in the transition from this zone to the zone of reaction products. Besides this assumption, we also assume that the reaction is of the first order with respect to the fluid reagent, the surface of the particle is equally accessible to it, and the diffusion obeys Fick's law.

Under these conditions, until the crust of the solid product is formed on the surface of the spherical particle (i.e., when the diffusion constant is constant in the volume of the particle), the boundary-value problem for the non-steady-state diffusion can be written as follows:

$$\frac{\partial c}{\partial \text{Fo}} = \frac{\partial^2 c}{\partial r^2} + \frac{2}{r}\frac{\partial c}{\partial r} - Kc, \tag{1}$$

$$c(r, 0) = 0, \ \text{Fo} = 0, \ 0 \leqslant r \leqslant 1, \tag{2}$$

$$\frac{\partial c}{\partial r} = 0, \ r = 0, \ \text{Fo} > 0, \tag{3}$$

$$\frac{\partial c}{\partial r} = \text{Bi}(1 - c), \ r = 1, \ \text{Fo} > 0, \tag{4}$$

where $c = C/C_0$; $r = R/R_0$; $\text{Fo} = D_0\tau/R_0^2$; $\text{Bi} = \beta R_0/D_0$; $K = K_0 R_0^2/D_0$.

$$c(r, \text{Fo}) = \frac{\text{Bi} \, \text{sh} \sqrt{K} r}{r[\sqrt{K}\,\text{ch}\sqrt{K} + (\text{Bi} - 1)\,\text{sh}\sqrt{K}]} + \sum_{1}^{\infty} \frac{2\,\text{Bi}\,\mu_n \sin\mu_n r \exp[-(K + \mu_n^2)\,\text{Fo}]}{r(K + \mu_n^2)(\text{Bi}\cos\mu_n - \mu_n \sin\mu_n)}, \tag{5}$$

where μ_n are the roots of the transcendental equation

$$\operatorname{tg} \mu = \frac{\mu}{1 - \mathrm{Bi}} .$$ (6)

According to our assumption that the reaction rate in an elementary volume of the particle depends directly on the concentration of the gaseous reagent in the given point, we obtain the following relations for the rate $G(\tau)$ and degree $\varepsilon(\tau)$ of chemical transformation of the particle:

$$G(\tau) = 4\pi h K_0 \int_0^{R_0} R^2 C(R, \tau)\, dR$$

or, in the dimensionless form,

$$j = \frac{G(\tau)}{4\pi C_0 h R_0 D_0} = K \int_0^1 r^2 c(r, \mathrm{Fo})\, dr,$$ (7)

$$\varepsilon = \frac{3}{4\pi R_0^3 \rho} \int_0^\tau G(\tau)\, d\tau = \frac{3 C_0 h K}{\rho} \int_0^{\mathrm{Fo}} d\mathrm{Fo} \int_0^1 r^2 c(r, \mathrm{Fo})\, dr.$$ (8)

Using these relations and expression (5) which describes the concentration distribution of the gaseous reagent in the volume of the particle, we obtain the following equations for $G(\tau)$ and $\varepsilon(\tau)$:

$$j = \frac{\mathrm{Bi}\,(K \operatorname{cth} \sqrt{K} - 1)}{\sqrt{K} \operatorname{cth} \sqrt{K} + \mathrm{Bi} - 1} + \sum_1^\infty \frac{2K\,\mathrm{Bi}\,(\mu_n \cos \mu_n - \mu_n)\exp - (K + \mu_n^2)\,\mathrm{Fo}}{(\mu_n \sin \mu_n - \mathrm{Bi} \cos \mu_n)(K + \mu_n^2)\,\mu_n} ,$$ (7')

$$\varepsilon = \frac{3K\,\mathrm{Bi}}{M} \left\{ \frac{(\sqrt{K} \operatorname{cth} \sqrt{K} - 1)\,\mathrm{Fo}}{K(\sqrt{K} \operatorname{cth} \sqrt{K} + \mathrm{Bi} - 1)} + \sum_1^\infty \frac{2(\mu_n \cos \mu_n - \sin \mu_n)[1 - \exp - (K + \mu_n^2)\,\mathrm{Fo}]}{\mu_n(\mu_n \sin \mu_n - \mathrm{Bi} \cos \mu_n)(K + \mu_n^2)^2} \right\},$$ (8')

where $M = \rho/c_0 h$.

Equations (7') and (8') are valid as long as there is no crust of the solid reaction product on the surface of the particle. To determine the moment of time $\mathrm{Fo}_0^* = \tau^* D_0 / R_0^2$ when the crust is formed, we construct an equation for the balance of matter in an elementary volume on the surface of the particle. Denoting the instantaneous density of the solid reagent in this volume by $q(R_0, \tau)$, we can write

$$dq(R_0, \tau) = - C(R_0, \tau) K_0 h\, d\tau.$$ (9)

Integrating (9) with respect to time and noting that for $\tau = 0$, we have $q(R_0, 0) = \rho$ and $\tau = \tau^*$ $q(R_0, \tau^*) = 0$, we obtain the following relation:

$$\int_0^{\tau^*} K_0 h C(R_0, \tau)\, d\tau = \rho \quad \text{or} \quad \int_0^{\mathrm{Fo}^*} K c(1, \mathrm{Fo})\, d\mathrm{Fo} = M.$$ (9')

Using Eq. (5) for $r = 1$ and in relation (9'), analyzing the values of the terms in the series and integrating (9'), we obtain the estimate:

$$\frac{M\,[\sqrt{K} \operatorname{cth} \sqrt{K} + \mathrm{Bi} - 1]}{K\,\mathrm{Bi}} < \mathrm{Fo}^* < \frac{M\,[\sqrt{K} \operatorname{cth} \sqrt{K} + \mathrm{Bi} - 1]}{K\,\mathrm{Bi}} \left[1 + \frac{100}{M} \right].$$ (10)

If the fluid reagent is gas, the value of the complex M is sufficiently large. It follows from (10) that for $M > 10^4$ (which is valid for the majority of solid-gas reactions), the moment when the crust of the solid reagent appears on the surface is determined, with accuracy up to 1%, by the following equation:

$$\mathrm{Fo}^* = \frac{M\,(\sqrt{K} \operatorname{cth} \sqrt{K} + \mathrm{Bi} - 1)}{K\,\mathrm{Bi}} .$$ (11)

Equations (7') and (8') are not suitable for engineering calculations. An analysis shows that, starting from some value Fo',

$$\mathrm{Fo}' = \frac{120\,(\sqrt{K}\,\mathrm{cth}\,\sqrt{K} + \mathrm{Bi} - 1)}{\mathrm{Bi}\,(\sqrt{K}\,\mathrm{cth}\,\sqrt{K} - 1)}\,, \tag{12}$$

the infinite series in these equations can be omitted since their contribution does not exceed 1%. A comparison of (11) and (12) shows that if $120K/(\sqrt{K}\,\mathrm{cth}\,\sqrt{K} - 1) < M$ (e.g., $K < 6800$ for $M = 10^4$), the ratio $\mathrm{Fo}'/\mathrm{Fo}^* < 1$. This means that before the crust is formed on the surface of the particle, the sum of the series in expression (8') becomes negligibly small and can be omitted. This makes it possible to determine the total degree of chemical transformation of the particle at time $\mathrm{Fo} = \mathrm{Fo}^*$ from the simple equation

$$\varepsilon^* = \frac{3\,(\sqrt{K}\,\mathrm{cth}\,\sqrt{K} - 1)}{K}\,. \tag{13}$$

It follows from expression (13) that $\varepsilon^* \to 1$ for $K \to 0$ and $\varepsilon^* \to 0$ for $K \to \infty$. This means that for large values of the combination $K_0 R_0^2 / D_0$ (practically for $K > 10^4$), the reaction penetration depth is very small and the reaction proceeds practically at the surface, where the crust is formed immediately.

In a sense the ratio $\mathrm{Fo}'/\mathrm{Fo}^* = 120K/M(\sqrt{K}\,\mathrm{cth}\,\sqrt{K} - 1)$ characterizes the time dependence of ε since for $\mathrm{Fo} > \mathrm{Fo}'$, this dependence is linear (in agreement with (8') and the above analysis), and Eq. (8') itself is valid only for $\mathrm{Fo} < \mathrm{Fo}^*$. For example, for the values $K < 100$ and $M > 10,000$ we have $\mathrm{Fo}'/\mathrm{Fo}^* = 0.044$ and, consequently, the graph of the function $\varepsilon = \varepsilon(\mathrm{Fo})$ has a linear character practically from the beginning of the reaction ($\varepsilon = 0.0118$) until the moment of formation of the crust at the surface.

As we noted above, after the formation of crust of completely transformed solid reagent at the surface, the diffusion conditions of the gas in the volume of the particle are changed since the gas does not react with the crust. In addition, the gas diffusion constant in the crust can differ from the original value.

By virtue of the above assumptions, the mathematical model of the reaction when a crust of inert material exists can be written as follows:

$$\frac{\partial c}{\partial \mathrm{Fo}} = m\left[\frac{\partial^2 c}{\partial r^2} + \frac{2}{r}\,\frac{\partial c}{\partial r}\right] - Kc, \tag{14}$$

$$\begin{cases} 0 \leqslant r < X(\mathrm{Fo}), & m = 1, \ K = K_0 R_0^2 / D_0, \\ X(\mathrm{Fo}) \leqslant r \leqslant 1, & m = D_1/D_0, \ K = 0, \end{cases} \tag{15}$$

$$\begin{cases} \dfrac{\partial c}{\partial r} = \dfrac{\mathrm{Bi}}{m}\,[1 - c], \ r = 1, \ \mathrm{Fo} > 0, \ m = \begin{cases} 1 & \text{for} \ X = 1, \\ D_1/D_0 & \text{for} \ X < 1, \end{cases} \\ \dfrac{\partial c}{\partial r} = 0, \ r = 0, \ \mathrm{Fo} > 1, \end{cases} \tag{16}$$

$$c \equiv 0, \ 0 \leqslant r \leqslant 1, \ \mathrm{Fo} = 0. \tag{17}$$

The appearance of an additional condition in Eq. (16) is due to the change of the diffusion constant of the gaseous reagent in the particle from value D_0 to D_1 when the dividing boundary $X(\mathrm{Fo})$ between the materials before and after the reaction is displaced from the surface of the particle inside the volume.

To solve the boundary-value problem (14)-(17), it is necessary to have an equation which determines the position of $X(\mathrm{Fo})$. The radius of this spherical boundary can be found from the integral balance relation.

$$\int_0^{\mathrm{Fo}(X)} Kc(X, \mathrm{Fo})\,d\mathrm{Fo} = M, \tag{18}$$

where $\mathrm{Fo}(X)$ is the moment of time in which the chemical transformation takes place at point with coordinate X.

The system of equations (14)-(18) was solved numerically by using an implicit scheme on nonuniform nets. We used here a driving method, taking into account the appropriate boundary conditions. The algorithm of the solution was realized on a BÉSM-6 computer.

The output parameters of the program of solution were the concentration fields of the gaseous reagent, the position of the dividing boundary X(Fo), the dimensionless reaction rate j(Fo), and the degree of chemical transformation of the particle ε(Fo). The values of the starting parameters K, Bi, m, and M were varied within the following wide limits: $1 \leqslant Bi \leqslant 1000$; $1 \leqslant K \leqslant 5000$; $0.01 \leqslant m \leqslant 100$; $10^3 \leqslant M \leqslant 2 \cdot 10^4$.

In all variants under study, the calculated values of the quantities ε^* and Fo^* agreed to a high accuracy with Eqs. (11) and (13). In addition, it was established that the reaction rate, the degree of chemical transformation, and the concentration fields depend practically only on the combination Fo/M, and not on each of these variables separately.

In Fig. 1 we show the time dependence of the degree of chemical transformation of the particle for Bi = 10, m = 1, and various values of the combination K. It follows from these graphs that the time from the moment when the crust appears on the surface to the completion of the chemical transformation depends weakly on the quantity $K = K_0 R_0^2 / D_0$.

An analogous pattern is observed also for other values of Bi and m. Physically, this is understandable. For small values of K, the reaction proceeds slowly. This is the reason why the crust is formed late (the degree of transformation is sufficiently large). The concentration of the gaseous reagent in the volume of the particle is in this case sufficiently large (Fig. 2). For large values of K, on the other hand, the crust is formed early and the reaction proceeds intensely. The time of its completion is therefore the same as in the first case.

For given values of K the time from the moment of formation of the crust to the full completion of the reaction depends strongly on Bi and m (Fig. 3). It is natural that for Bi → 0 or m → 0 it tends to infinity. It is seen from Fig. 3, which is constructed for K = 49, that for Bi > 100 and m > 100, the mass release on the surface and the diffusion through the crust of the reacted substance do not limit the rate of transformation of the solid reagent which is determined in this case only by the reaction in the core of the particle.

Information about the limiting stage of the process can be obtained also from the concentration fields of the gaseous reagent in the volume of the particle. For example, it follows from Fig. 2 that for one set of values of K = 49, Bi = 500, and m = 100 (curve 7), the reaction rate is determined only by the rate of transformation in the core of the particle. For Bi = 10 and m = 0.01 (curve 8), however, the reaction rate is determined by the diffusion through the crust of the solid product, and for Bi = 1, m = 100, and K = 1000, mainly by the intensity of the mass transfer from the surrounding medium to the surface of the particle.

Thus, it follows from Figs. 2 and 3 that the effect of the chemical reaction on the rate of formation of the solid product is different for different values of Bi and m. For one particular value of K, the chemical reaction can either slow down the process or not limit at all the rate of formation of the reaction products.

An analysis of the results of the calculation showed that the thickness of the reaction zone δ under the crust of the reacted matter depends only on the combination $K = K_0 R_0^2 / D_0$. It can be estimated as

$$\delta = X - \sqrt[3]{1 - \varepsilon}, \tag{19}$$

where the quantity $\sqrt[3]{1 - \varepsilon}$ is the radius of some fictitious sphere which would be formed if all the unreacted solid reagent was concentrated in its volume.

It is seen from Fig. 4 that the thickness of the reaction zone decreases with increasing K and tends to zero as K → ∞. Practically, already for K = 5000 when the reaction depth is $0.015 R_0$, one can assume that the reaction proceeds mainly on the dividing boundary between the solid phases of the original reagent and the reaction product. We found the following empirical formula for the thickness of the reaction zone:

$$\delta = X \left(1 - \sqrt[3]{1 - \frac{3(\sqrt{K}\, X \operatorname{cth} \sqrt{K}\, X - 1)}{K X^2}} \right), \tag{20}$$

which describes, with accuracy up to 1%, the results shown in Fig. 4.

The results of the numerical solution of the problem (14)-(18) for the calculation of processes and instruments for the thermochemical processing of a multicomponent polydispersed granular material are not always useful since the complicated procedure for the calculation
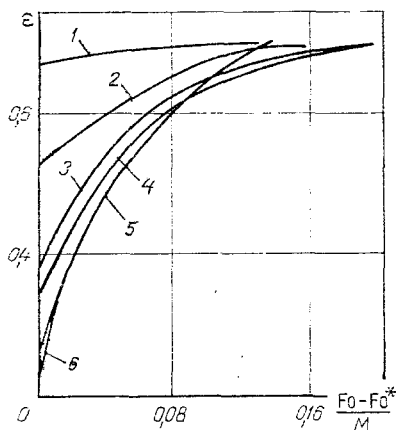
Fig. 1   Fig. 2

Fig. 1. Change of the degree of chemical transformation with time after the appearance of the crust on the surface of the particle for Bi = 10, m = 1, and various K. Curves 1, 2, 3, 4, 5, and 6 correspond to K = 1, 10, 49, 100, 1000, and 5000, respectively.
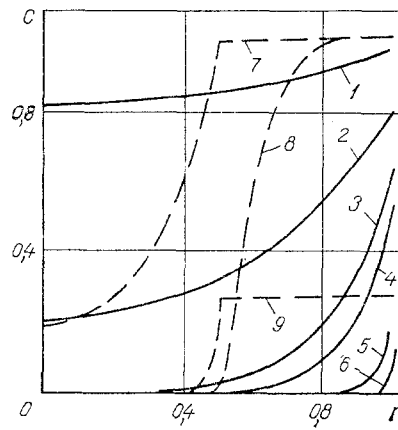
Fig. 2. Concentration field of the gaseous reagent in the volume of the particle. Curves 1, 2, 3, 4, 5, and 6 correspond to X = 1, Bi = 10, and K = 1, 10, 49, 100, 1000 and 5000, respectively; 7) X = 0.5, m = 100, Bi = 500, K = 49; 8) X = 0.5, m = 0.01, Bi = 10, K = 49; 9) X = 0.5, m = 100, Bi = 1, K = 1000.

of the degree of transformation and the reaction rate must be carried out many times for different dimensions and state of the composition of the particles. The program for the calculation of the thermochemical transformation process is in this case hindered also by the fact that the system of these equations must be solved simultaneously with other equations which describe the heat and mass exchange processes in the working volume of the instrument. It is therefore more convenient to use approximate analytical formulas, corrected by taking into account the results of the numerical solution.

To obtain an analytical expression which reflects the time dependence of the degree of chemical transformation, one can use the solution of the present problem in the quasistationary approximation [1]. This approach is justified by the fact that the mass of the gaseous reagent in the pores of the particle is negligible in comparison with the mass of the solid reagent. The change of concentration fields of the gas in the reaction process takes place much faster than the motion of the reaction front, i.e., these fields practically do not differ from stationary ones.

Kirakosyan [1] obtained the functional dependence X = X(Fo) under the condition that $X\sqrt{K} > 3$. Below, we have succeeded in solving this problem without this limitation. We obtained the expression

$$\frac{\text{Fo} - \text{Fo}^*}{M} = \frac{1}{m}\left[\frac{1 - X^2}{2} - \frac{1 - X^3}{3}\right] + \frac{1 - X^3}{3\text{Bi}} + \frac{1}{K}\ln\frac{\text{sh}\,\sqrt{K}\,(\sqrt{K}\,\text{cth}\,\sqrt{K} - 1)}{\text{sh}\,\sqrt{K}\,X(\sqrt{K}\,X\,\text{cth}\,\sqrt{K}\,X - 1)}. \quad (21)$$

Calculations showed that if the equation

$$\varepsilon = 1 - X^3\left[1 - \frac{3(\sqrt{K}\,X\,\text{cth}\,\sqrt{K}\,X - 1)}{KX^2}\right] \quad (22)$$

is used to determine the degree of transformation [this equation is obtained from (19) and (20)], the difference between (21) and (22) and the results of the mathematical model [Eqs. (14)-(18)] for the reaction does not exceed 10%.

To improve the approximate method for the calculation of the reaction rate and the degree of chemical transformation still further, the form of the starting equations (21) and (22) was changed somewhat. Calculations showed that if the dependence ε = ε(X) is represented as
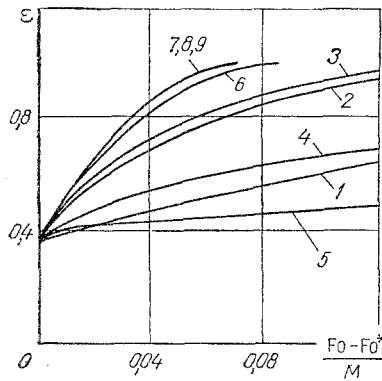
Fig. 3

Fig. 4

Fig. 3. Change of the degree of chemical transformation after formation of the crust on the surface of the particle for K = 49 and different Bi and m: 1, 2, 3) m = 1, Bi = 1, 10, 100; 4, 5) Bi = 10, m = 0.1, 0.01; 6, 7) Bi = 100, m = 10, 100; 8, 9) Bi = 1000, m = 100, 1000.
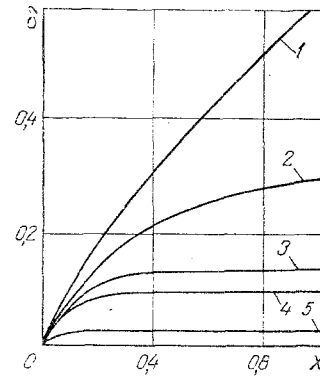
Fig. 4. Dependence of the thickness of the reaction zone on the position of the reaction front and the value of K. Curves 1, 2, 3, 4, and 5 correspond to K = 1, 10, 49, 100, and 1000.

$$\varepsilon = 1 - X^3 \left\{ 1 - \left[\frac{3(\sqrt{K}\, X \operatorname{cth} \sqrt{K}\, X - 1)}{KX^2} \right]^{X^{1,5}} \right\},$$

(23)

and X(Fo) is represented by the equation

$$\frac{Fo - Fo^*}{M} = \frac{1}{m} \left[\frac{1 - X^2}{2} - \frac{1 - X^3}{3} \right] + \frac{1 - X^3}{3Bi} + \frac{1}{K} \ln \frac{X^3 \operatorname{sh} \sqrt{K} (\sqrt{K} \operatorname{cth} \sqrt{K} - 1)}{\operatorname{sh} \sqrt{K}\, X (\sqrt{K}\, X \operatorname{cth} \sqrt{K}\, X - 1)},$$

(24)

the maximum deviation from the results obtained from the mathematical model (14)-(18) is 6% for the dependence $\varepsilon = \varepsilon(Fo)$, and 8% for $j = j(Fo)$. The function $j(X)$ is here calculated using the equation

$$j = \frac{m\, Bi\, X\, (\sqrt{K}\, X \operatorname{cth} \sqrt{K}\, X - 1)}{m\, Bi + [Bi\,(1 - X) + mX]\, (\sqrt{K}\, X \operatorname{cth} \sqrt{K}\, X - 1)}.$$

(25)

It should be noted that this deviation is observed for variation of the starting parameters in very wide limits and, in addition, for large ε (larger than 0.8). Thus, Eqs. (23) and (25), supplemented by (24), can be recommended for calculating the reaction rate and degree of chemical transformation of the particle after formation of the crust of solid product on the surface of the particle.

An analysis of the obtained dependences for $\varepsilon(Fo)$ showed that, in particular cases, they can be used to obtain the corresponding equations proposed by various authors [2-6] for the description of the kinetics of heterogeneous reactions. For example, for large values of K (practically for K > 5000) one can assume [see (11) and (13)] that, starting from the very beginning of the reaction, a crust of solid product is formed on the surface of the particle. Consequently, in this case, (24) can be used from the moment Fo = 0. For the above values of K, the reaction proceeds practically only on the boundary between the solid product and solid reagent, and the dependence $\varepsilon = \varepsilon(X)$ can be simplified [see Eq. (23)]:

$$\varepsilon = 1 - X^3.$$

(26)

In addition, calculations show that in this case, the last term on the right-hand side of expression (24) can be replaced by $(1 - \overline{X})/\sqrt{K}$, with an error of no more than 7%. Thus, after some transformations, Eq. (24) yields the equation

$$\tau = \frac{\rho R_0}{C_0 h} \left[\frac{1 - X}{K_n} + \left(\frac{1 - X^2}{2} - \frac{1 - X^3}{3} \right) \frac{R_0}{D_1} + \frac{1 - X^3}{3\beta} \right].$$

(27)

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Here $K_n = \sqrt{K_0 D_0}$.

A similar formula as (27) was proposed in [2-4] for the description of the kinetics of the reactions $solid_1 + gas_1 \to solid_2 + gas_2$ which proceed in a mixed (diffusion-kinetic) regime. It is easily verified that Eq. (27) yields the equation for diffusion kinetics [5] if $K_n \to \infty$, $\beta \to \infty$, and chemical kinetics [6] if $D_1 \to \infty$, $\beta \to \infty$.

The condition $K > 5000$ for which Eq. (27) is valid is conveniently written in the form $K_n R_0/D_1 > 70.7/m$. Hence, it follows that if $m = D_1/D_0 \gg 1$, (27) can be used in the calculations in the majority of practically interesting cases.

NOTATION

C_0 and C, concentrations of the gaseous reagent in the surrounding medium and in the volume of the particle, respectively; R_0 and R, radius of the particle and the radial coordinate; D_0 and D_1, diffusion constants of the gaseous reagent in the reacting solid substance and in the solid reaction product, respectively; K_0, reaction rate constant; h, stoichiometric coefficient; G and j, reaction rate and its dimensionless value; X, dimensionless radial coordinate of the reaction front; ρ, density of the solid reagent; ε, degree of chemical transformation; τ, instantaneous time; δ, dimensionless value of the thickness of the reaction zone; β, mass transfer coefficient from the gases to the surface of the particle.

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METHOD FOR INVESTIGATING HETEROGENEOUS PROCESSES IN A SOLID—SOLID SYSTEM IN MULTISECTIONED V-SHAPED CONTINUOUS ACTION APPARATUS

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The process of contact drying of thermolabile materials by adsorbents in V-shaped apparatus with periodic and continuous action is analyzed experimentally and theoretically.

There exists a wide class of thermolabile materials for which thermal drying is inapplicable. For many of these materials, the method of contact drying by adsorbents is effective [1, 2]. However, up to the present time, under commercial conditions, such processes are mainly carried out in continuous action mixers of the drum type (V-shaped, biconical, tetrahedral, etc. [3]), which have all the disadvantages of periodic action apparatus and, in addition, can lead to considerable polarization and erosion of the particles depending on the physical properties of the material being worked. These disadvantages are partially absent